Lipid/Polyelectrolyte Complexes – Effects of the Polyelectrolyte Architecture on the Self-assembled Structures

Supporting Information

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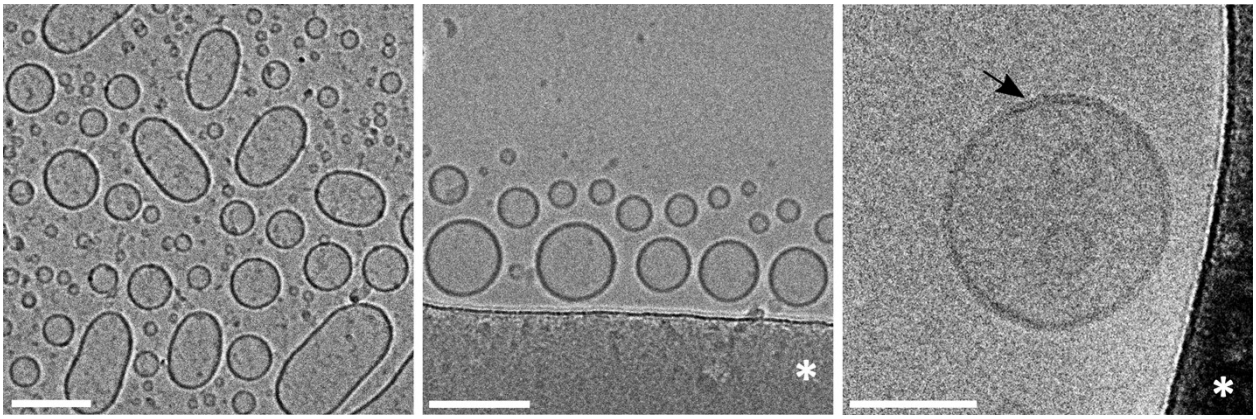

Figure S1. Cryo-TEM images of pure DOTAP (c = 2 mM), showing unilamellar liposomes of various sizes. The lipid double layer is resolved in the right panel (black arrow). The large dark domains are parts of the perforated carbon film (white asterisks). Scale bars are 100 nm, except for right panel where the scale bar equals 50 nm.

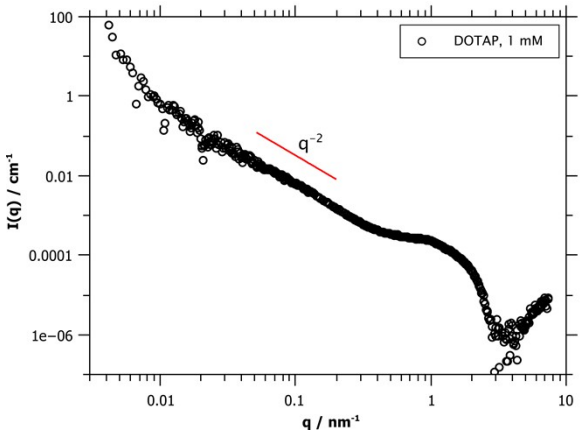

Figure S2. SAXS data of pure DOTAP (c = 1 mM), showing unilamellar liposomes of various sizes.

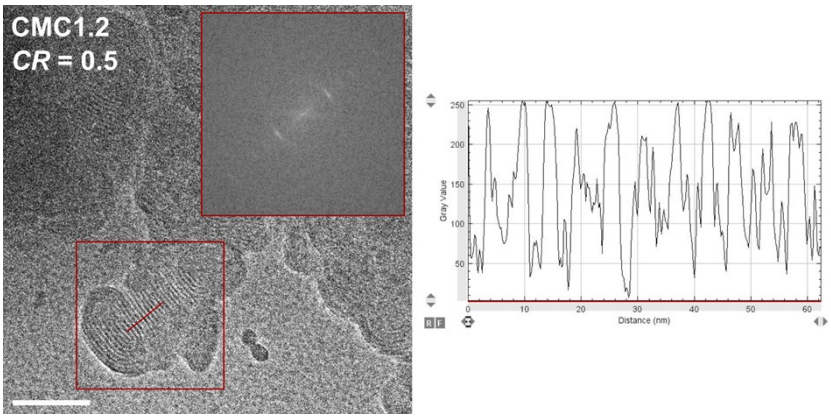

Figure S3. Exemplary Fast Fourier Transform (FFT), showing the lamellar order of a DOTAP/CMC1.2, *CR* = 0.5 complex and profile plot used to deduce the lamellar spacing from the microscopy images.

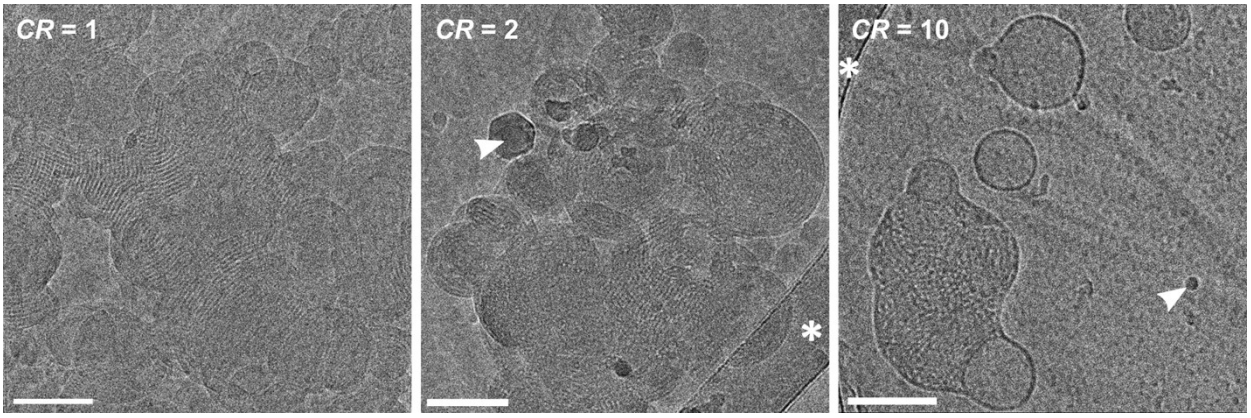

Figure S4. Cryo-TEM images of 1 mM DOTAP mixed with CMC1.2 at different charge ratios (*CR* = 1, 2, 10), all on the side of DOTAP excess. At very high charge ratios DOTAP liposomes can be seen coexisting with multilayered mixed complexes. The large dark domains are parts of the perforated carbon film (white asterisks), dark spots are ice crystals (white arrowheads). Scale bars are 100 nm.

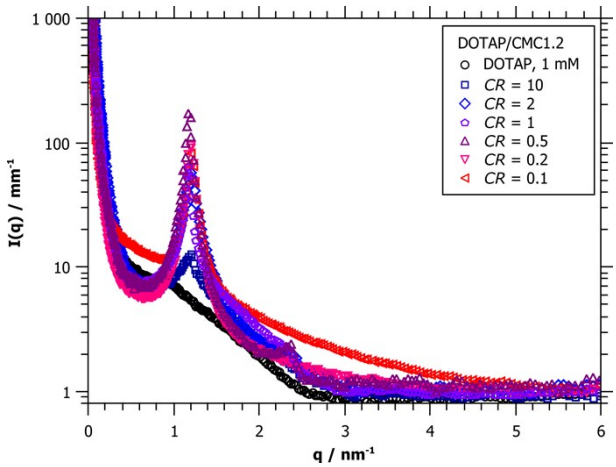

Figure S5. SAXS data of the series DOTAP/CMC1.2, *c*(DOTAP) = 1 mM, at different *CR*. The data was normalized for the incoherent background at high *q* so that the relative peak heights become visible. The most pronounced peak, hence the sample with the highest order was found at *CR* = 0.5.

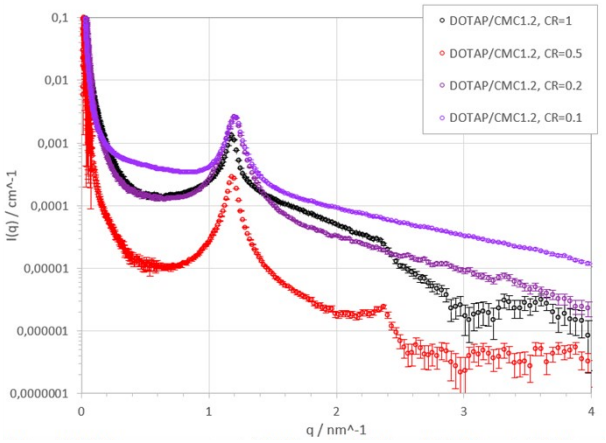

Figure S6. SAXS data of the series DOTAP/CMC1.2, *c*(DOTAP) = 1 mM, at different CR, including errors. In most cases, the error bars are smaller than the shown symbols and all peaks, including higher order peaks are significant compared to the error. Thus, errors are omitted for more clarity in other SAXS graphs.

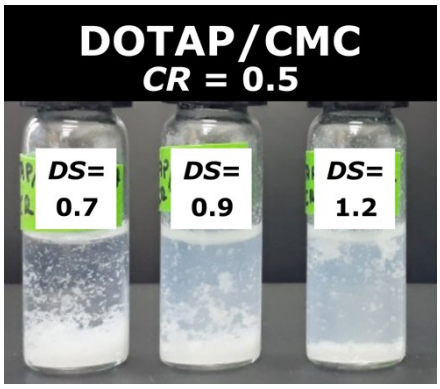

Figure S7. A photo of DOTAP/CMC samples at *CR* = 0.5, *c*(DOTAP) = 1 mM, at different charge densities, defined by the degree of substitution of the CMC, *DS* = 0.7, 0.9, and 1.2. The amount of precipitation decreased with increasing charge density, while the aggregate size in the supernatant is increasing (as seen by the color).

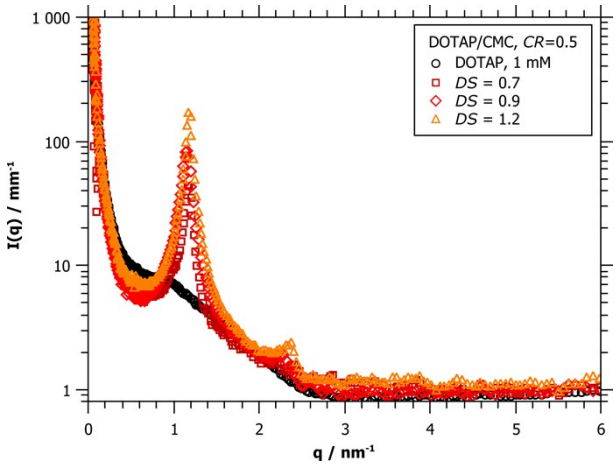

Figure S8. SAXS data of 1 mM DOTAP mixed with CMC of different charge densities, *DS* = 0.7, 0,9, and 1.2, all at *CR* = 0.5. The data was normalized for the incoherent background at high *q* so that the relative peak heights become visible. The 1st and 2nd order peaks both increase with higher charge density, indicating an increased order of the multilayer structures within the aggregates.

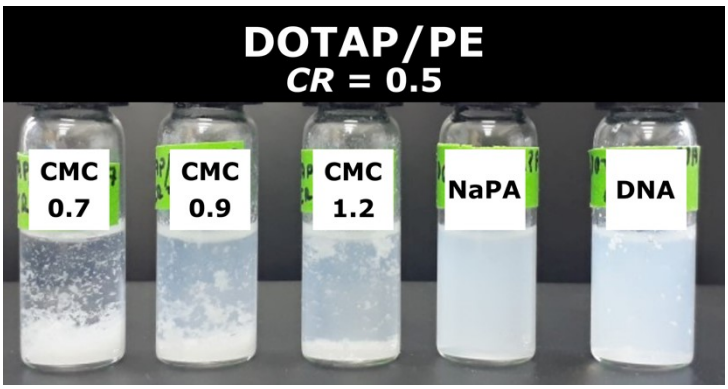

Figure S9. Photo of samples of DOTAP mixed with different polyelectrolytes, all at *CR* = 0.5, *c*(DOTAP) = 1 mM.

Table S1. Summary of peak positions of all samples and corresponding lamellar spacing deduced from SAXS measurements.

	1st peak / nm^{-1}	**2nd peak / nm^{-1}**	**spacing / nm**
DOTAP, 1mM	-	-	-
DOTAP/CMC0.7, *CR*=0.5	1.16	-	5.39
DOTAP/CMC0.9, *CR*=0.5	1.16	2.29	5.39
DOTAP/CMC1.2, *CR*=0.5	1.18	2.37	5.33
DOTAP/NaPA, *CR*=0.5	1.27	2.53	4.95
DOTAP/DNA, *CR*=0.5	1.04	2.07	6.03
DOTAP/PSS, *CR*=0.5	1.36	-	4.62
DOTAP/CMC1.2, *CR*=10	1.20	-	5.25
DOTAP/CMC1.2, *CR*=2	1.21	2.40	5.17
DOTAP/CMC1.2, *CR*=1	1.18	2.32	5.33
DOTAP/CMC1.2, *CR*=0.2	1.20	-	5.25
DOTAP/CMC1.2, *CR*=0.1	1.21	-	5.17